Two-dimensional magnetic order in $Pb_2Sr_2TbCu_3O_8$

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Neutron diffraction techniques have been used to investigate the magnetic ordering of the Tb ions in polycrystalline $Pb_2Sr_2TbCu_3O_8$. Significant magnetic correlations are found to develop below ~10 K, and these correlations are two dimensional in nature, representative of the strongly anisotropic magnetic interactions in these layered materials. The correlation length increases with decreasing temperature, and long-range order is observed to develop at $T_N \approx 5.5$ K. However, the sawtooth Bragg profile for the scattering demonstrates that the long-range order is two dimensional, rather than three dimensional, with a spin configuration where nearest-neighbor spins are antiparallel. The scattering profile can be explained quantitatively by assuming long-range order within the *ab* plane, with no significant correlations along the *c* axis to the lowest temperatures measured (1.36 K). Our results suggest that the system is well described by a two-dimensional Ising model. The only other (pure) system where a crossover to three-dimensional behavior is *not* observed is $DyBa_2Cu_4O_8$.

One of the most remarkable properties of the Cu and $\mathcal{RE}$ ($\mathcal{RE}$=rare-earth elements) magnetism of the high T_c oxides is that they are two dimensional (2D) in nature. The 2D behavior of the Cu magnetism originates from the strong Cu-O bonding within the layers, which yields highly anisotropic exchange interactions between the Cu ions. On the other hand, the $\mathcal{RE}$ atoms form an orthorhombic sublattice where the nearest-neighbor distance along the *c* axis is much greater than that within the *ab* plane, which again leads to highly anisotropic magnetic interactions between the $\mathcal{RE}$ ions and generates the 2D magnetism. Such 2D behavior of the $\mathcal{RE}$ ions has been observed in the $\mathcal{RE}Ba_2Cu_3O_7$[1,2] ($\mathcal{RE}$1237), $\mathcal{RE}Ba_2Cu_4O_8$[3] ($\mathcal{RE}$1248), and $\mathcal{RE}_2Ba_4Cu_7O_{15}$[4] ($\mathcal{RE}$24715) systems. Among them, a rod of scattering characteristic of 2D behavior has been observed[2] in single crystals of Er1237 and Dy1237, and their order parameters are found to obey the Onsager[5] solution for the $S=\frac{1}{2}$, 2D Ising antiferromagnet. Moreover, for the Dy ions in the Dy1248 a geometric cancellation of the already weak interactions occurs along the *c* axis, which renders the system two-dimensionally ordered even at temperatures well below the Néel temperature.[3]

In this article we report another system which exhibits 2D magnetic order, as observed by neutron diffraction experiments. In this material we observe significant 2D magnetic correlations of the Tb spins in the $Pb_2Sr_2TbCu_3O_8$ (Tb22138) compound below $T \sim 10$ K. Long-range order is observed to develop at $T_N \approx 5.5$ K, but even at $T=1.36$ K the magnetic correlations are still mainly two dimensional, with nearest-neighbor spins within the *ab* plane being aligned antiparallel.

The $\mathcal{RE}$22138 systems are even more anisotropic in their physical properties than those of the $\mathcal{RE}$1237 system,[6,7] as there are more intervening layers of atoms stacked along the *c* axis. The rare-earth atoms in the $\mathcal{RE}$22138 system form an orthorhombic unit cell where the distance between the nearest neighbors along the *c* axis is more than four times that in the *ab* plane. It is clear that the crystallographic anisotropy naturally leads to highly anisotropic magnetic interactions. The rare-earth unit cell is related to the nuclear unit cell of the compound by rotation of the *a* and *b* axes of the nuclear unit cell by ~45° and reducing their lengths by $\sqrt{2}$. We employ the notation of (abc) and $(a'b'c')$ to represent the axes of the nuclear and rare-earth unit cells, respectively. We note that by using this system of notation, the *ab* plane and $a'b'$ plane are essentially the same planes.

Powder samples of $Pb_2Sr_2TbCu_3O_8$ were prepared by the standard solid-state reaction techniques, and the details of the sample preparation technique can be found elsewhere.[8] Both x-ray and neutron diffraction were used to characterize the sample. The x-ray diffraction pattern is well described by the orthorhombic Y22138 structure,[6] which is only slightly deviated from tetragonal symmetry, with room-temperature lattice parameters $a=5.452(7)$ Å, $b=5.459(9)$ Å, and $c=15.66(2)$ Å for the chemical unit cell of the compound. The nearest-neighbor distances of the Tb ions are, therefore, $a' \approx b' = 3.856$ Å within the $a'b'$ plane and $c' = 15.66$ Å along the c' axis, respectively.

Neutron diffraction measurements were performed at the Research Reactor at the U.S. National Institute of Standards and Technology. The data were collected using the BT-9 triple-axis spectrometer which was operated in double-axis mode. A pyrolytic graphite PG(002) crystal was used as the monochromator to extract neutrons of energy 14.8 meV ($\lambda = 2.351$ Å). A PG filter was also placed after the monochromator position to suppress higher-order wavelength contaminations. The angular collimations used were 40′ before the monochromator, and 48′-48′ before and after the sample, respectively. The sample was mounted in a cylindrical aluminum can filled with helium exchange gas to facilitate thermal conduction at low temperatures. A pumped ^{4}He cryostat was used to cool the sample, and the lowest temperature

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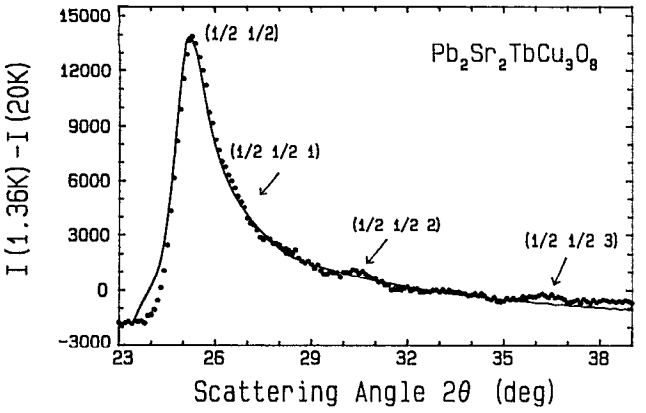

FIG. 1. Magnetic intensities observed in $Pb_2Sr_2TbCu_3O_8$ at $T=1.36$ K, which is well below the ordering temperature. The pronounced asymmetry of the observed peak is a classic profile of the two-dimensionally ordered system. The magnetic peaks may be indexed as the $(\frac{1}{2}\frac{1}{2})$ rod of scattering based on the Tb unit cell. Small 3D-like Bragg peaks, with positions that may be indexed as the $\{\frac{1}{2}\frac{1}{2}1\}$, $\{\frac{1}{2}\frac{1}{2}2\}$, and $\{\frac{1}{2}\frac{1}{2}3\}$ reflections, are also revealed. The solid line is a fit to the 2D theoretical profile.

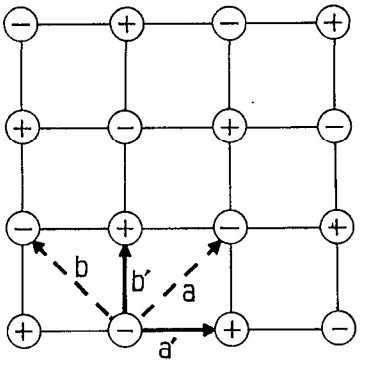

FIG. 2. The 2D spin configurations of the Tb ions within the $a'b'$ plane. The nearest-neighbor spins are aligned antiparallel, and the plus and minus signs then represent spins that are in opposite directions. a' and b' indicate the axes of the Tb unit cell, and a and b indicate the axes of the nuclear unit cell of the compound.

obtained was 1.36 K. For each temperature change, sufficient time was given to allow the sample to reach thermal equilibrium.

Three powder diffraction sets of data were collected over the range of scattering angles from $2\theta=5°$ to 65°: One at a temperature well below, one just above, and the other well above the Néel temperature. The subtraction technique[9] was used to isolate the magnetic signal from the nuclear one, where the diffraction pattern taken at a temperature well above the ordering temperature was subtracted from the one taken at low temperature.

The magnetic diffraction pattern obtained at $T=1.36$ K is shown in Fig. 1, where the diffraction pattern taken at $T=20$ K serves as the nonmagnetic "background" has been subtracted from the data. The sawtooth Bragg profile of the observed peak is a classic profile of a two-dimensionally ordered system.[3,10] The scattering pattern is dominated by a single 2D-like peak, even though we were well below the Néel temperature ($T_N\approx5.5$ K, see below). The solid curve in the figure is a least-squares fit to the theoretical 2D scattering profile convoluted with the Gaussian instrumental resolution function, assuming long-range order within the $a'b'$ plane and no correlations along the c'-axis direction. Based on the Tb unit cell, the observed asymmetrical peak corresponds to the $(\frac{1}{2}\frac{1}{2})$ rod of scattering. This is the same type of scattering that has been observed[3] in Dy1248. The magnetic correlations along the c'-axis direction are much weaker than those within the $a'b'$ plane. However, the observed intensities shown in Fig. 1 also suggest some 3D ordering character, where small 3D peaks at $2\theta=26.23°$, 30.09°, and 36.23°, which would correspond to the $\{\frac{1}{2}\frac{1}{2}1\}$, $\{\frac{1}{2}\frac{1}{2}2\}$, and $\{\frac{1}{2}\frac{1}{2}3\}$ Bragg reflections, respectively, are evident. This may indicate that a small fraction of the samples is ordered three dimensionally at low temperatures.

The presence of the $(\frac{1}{2}\frac{1}{2})$ rod of scattering demonstrates that the 2D magnetic unit cell of the Tb ions is doubled in size compared to the nuclear one along both the a' and b' axes. The nearest-neighbor spins of the Tb ions are therefore aligned antiparallel. The 2D spin configuration for the Tb moments in the $a'b'$ plane is shown in Fig. 2, where the a and b axes of the nuclear unit cell of the compound are also indicated. In addition, the weak $(\frac{1}{2}\frac{1}{2}l)$ type of reflections, with $l=$integer, revealed in Fig. 1 indicate that the Tb spins have the tendency to align parallel along the c' axis. These data suggest that 3D order may set in at lower temperatures. Further work at lower temperatures is planned to clarify this point.

The low-temperature ordered moment was obtained by comparing the magnetic to nuclear intensities. For the case of 2D magnetic ordering, we need to integrate the magnetic scattering over the angular range corresponding to the first Brillouin zone. Comparing the magnetic intensity with the {001} nuclear intensity establishes that the saturated moment for each Tb ion is $\langle\mu_Z\rangle=7.43\pm0.02\ \mu_B$, assuming the spin direction is along the c'-axis direction. This choice of the c'-axis direction is consistent with the moment direction usually found in these materials.

The temperature dependence of the intensity at the 2D $(\frac{1}{2}\frac{1}{2})$ peak position is shown in Fig. 3. The magnetic Bragg intensity is proportional to the square of the ordered moment, which is the order parameter of the phase transition. Assuming that the magnetic peak profile remains the same for the temperature range studied, a measurement of the temperature dependence of the peak intensity then measures the square of the order parameter. The sharp drop in intensity shown in Fig. 3 is typical for a 2D magnetic phase transition, while the rounding in the vicinity of the ordering temperature is typical for the (critical) scattering observed in a powder. The Néel temperature is estimated to be $T_N\approx5.5$ K.

Above the ordering temperature magnetic scattering was also observed, and the shape of this scattering also demonstrates the 2D magnetic character of the system. Figure 4 shows the $(\frac{1}{2}\frac{1}{2})$ rod of scattering observed at $T=6$ K, which is just above the ordering temperature. The intensity on the lower angle side of the peak does not show as sharp an increase because of the finite correlation range above the ordering temperature. The solid curve is a fit to the theoreti-

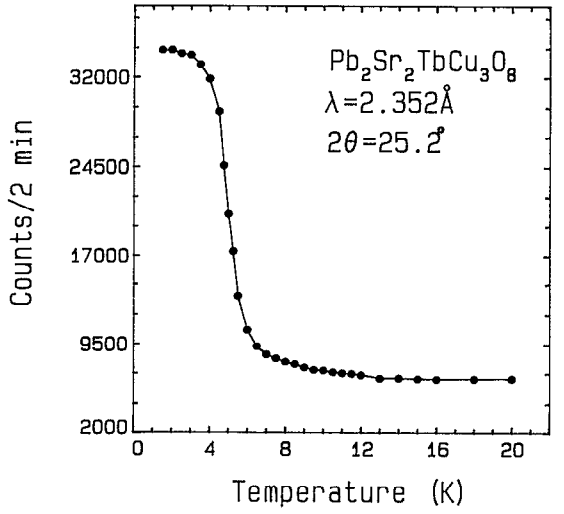

FIG. 3. Temperature dependence of the $(\frac{1}{2}\frac{1}{2})$ peak intensity, showing the variation of the square of the staggered magnetization with temperature. The sharp drop in intensity is typical for a 2D order parameter. The ordering temperature is determined to be $T_N \approx 5.5$ K. The solid curve is a guide to the eye only.

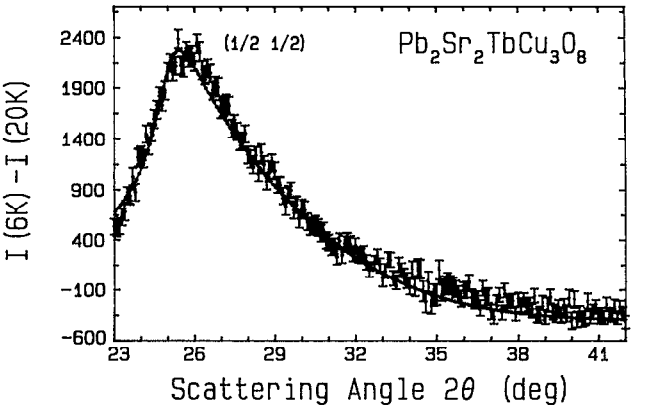

FIG. 4. The $(\frac{1}{2}\frac{1}{2})$ rod of scattering observed at $T=6$ K, which is just above the ordering temperature. The observation of this scattering directly demonstrates the 2D energetics of the system. The solid line is a fit to the 2D theoretical profile with a correlation length of $\xi=550$ Å within the $a'b'$ plane and no correlations along the c'-axis direction.

cal 2D profile, assuming a finite correlation length within the $a'b'$ plane and no correlations along the c'-axis direction. The 2D Bragg rod gives an excellent fit to the observed data. We, hence, conclude that there are strong magnetic correlations within the $a'b'$ plane while those along the c'-axis direction are weak, and the fundamental energetics is 2D in nature. The intrinsic linewidth of this peak is related to the inverse 2D correlation length, and the correlation length that we obtained from the fit at $T=6$ K is $\xi=550$ Å. Further work is in progress on the temperature dependence of the 2D magnetic correlation length in this system.

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